Ultrafast Optical Modulation using Single-crystal Films of Organic Nonlinear Optical Materials

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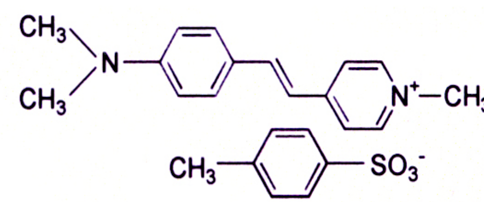
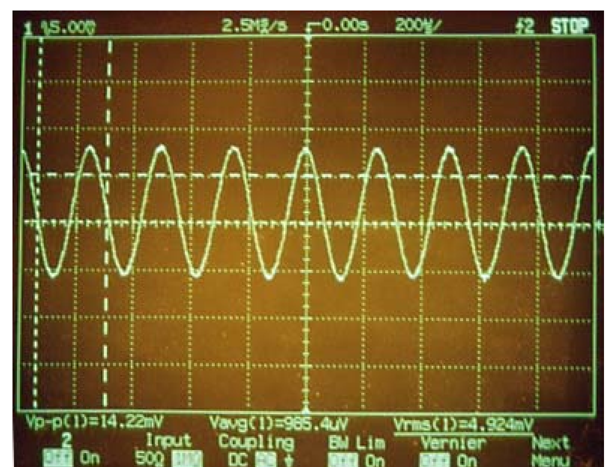
•The objective of this project is to prepare single-crystal films of unique organic materials and demonstrate exceptionally large electro-optic effects with ultrafast response.

• Single-crystal films of 4'-dimethylamino-N-methyl-stilbazolium tosylate (DAST) has been prepared on quartz substrate using "shear method". The shear method is the only method presently available for preparation of organic single crystal films. Films with areas more than 1 cm^2 are obtained. The DAST molecules are oriented close to parallel to the substrate in the single-crystal film. Electro-optic measurements at near resonant wavelengths have shown extremely large electro-optic coefficients and electro-absorption. The magnitude of the electro-optic coefficient (r_{11}) at 633 nm is 770 pm/V (more than 25 times that of lithium niobate). The measured electroabsorption that results from the imaginary part of the electro-optic effect is also large enough for use in modulation applications.



Thin single-crystal film of DAST on a quartz substrate. The film is about 1 cm^2 in area and 3 μm in thickness.



Electro-optic modulation at 633 nm in DAST film showing an electro-optic coefficient of 770 pm/V.

Recent Results:

Single-crystal films of DAST have been prepared using the "shear method". Growth at a higher temperature has increased the areas of the films. Electroabsorption and resonant electro-optic effect in these films have been measured at 633 nm and 488 nm. The absorption coefficients of the film at 488nm and 633 nm are ~ 6×10^3 cm^{-1} and ~ 10^3 cm^{-1} respectively. Gold electrodes were deposited on the film to apply the electric field along the dipole axis (a axis) which could be easily identified using the dichroism in the film. The beam propagated perpendicular to the film (0.3 μm thick) with the polarization oriented at an angle (θ) with respect to the dipole axis (a-axis). An ac field of about 1 V/μm (at 4 kHz) was applied along the dipole axis (a-axis). The transmitted beam was detected using a photodiode and recorded on an oscilloscope. In the measurement of the real part of the electro-optic coefficient, a Babinet–Soleil phase compensator was placed after the sample to compensate for the inherent birefringence. After the compensator, the beam passed through an analyzer (field-induced birefringence) before being detected by a photodiode. For the electroabsorption measurement, the compensator and the analyzer were removed. Modulation depths in electroabsorption of 1.4% at 633 nm and 3.1 % at 488nm were observed (film thickness 0.3 μm). The modulation depth at 488nm is larger than that at 633 nm since the absorption coefficient (α) at 488nm is larger than at 633nm. The oscilloscope trace of the electro-optic modulation at 633 nm for an applied ac field of 1 V/μm is shown in the attached figure. A modulation depth of 4.2% was observed. The real part of the electro-optic coefficient as determined from this measurement is r_{11} = 770 pm/V (about 25 times that of lithium niobate). The exceptionally large magnitude of the electro-optic coefficient at 633 nm is due to resonance enhancement.

The modulation signal for electroabsorption follows the applied electric field in time. This is different than the electroabsorption observed in conjugated polymers and multiple quantum wells in which cases the modulation depends quadratically on the electric field. In the present case, $\Delta\alpha$ depends linearly on electric field and therefore becomes positive and negative as the direction of the electric field is changed. More specifically, the change in absorption coefficient at any given field is given by $\Delta\alpha = -(\Delta T/T)/L$, where L is the thickness of the film. The modulation depths for various angles (θ) of polarization of the incident beam with respect to the dipole axis (a-axis) were measured. The magnitude of the modulation depth is the highest when the polarization is along the dipole axis and it decreases as the angle of polarization is increased. The magnitude of $\Delta\alpha$ is proportional to the imaginary part of the electro-optic coefficient r_{11}. The magnitudes of the imaginary parts of the electro-optic coefficient as determined are 104 pm/V and 259 pm/V at 633 nm and 488 nm respectively.

Electro-optic Fabry-Perot Modulator based on DAST Film:

Preliminary work on fabrication of an electro-optic Fabry-Perot modulator with a DAST film in the cavity has been completed. Initial results show a modulation depth of about 100% for a field of about 0.5 V/μm at 633 nm (film thickness ~ 8 μm). This is promising for practical applications.

STUDENTS: Two students (one M.S. and one Ph.D.) are being supported in this project.